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The connection between non-exponential relaxation and fragility in supercooled liquids

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Abstract

Among the outstanding problems in the theory of supercooled liquids are the reasons for the rapid increase in their viscosity and relaxation times as the temperature is lowered towards the glass transition temperature T_g , the non-exponential time dependence of the relaxation, and the possible connection between these two properties. The ferromagnetic Potts model on a square lattice is a simple system that is found to exhibit these properties. Our calculations show that in this system the connection between them is associated with the dependence on temperature and time of the average environment of the sites. Some of the consequences of this for understanding the behavior of supercooled liquids are discussed.

I. Introduction

There are a number of features common to most supercooled glass-forming liquids near their glass transition temperature T_g , and which differ appreciably from those of normal liquids and of crystalline solids.¹ Two of the most prominent of these are the temperature dependence of the properties close to T_g and their time dependence. In many supercooled liquids it is found that as the temperature is lowered towards T_g properties such as the viscosity η and the mean dielectric and mechanical relaxation times τ increase much more rapidly than the Arrhenius behavior associated with a fixed activation energy, which would give $\ln(\tau/\tau_0) = E/(kT)$. According to Angell's classification scheme,² strong liquids are those in which the departure from the Arrhenius behavior is slight and fragile ones are those for which the temperature dependence departs strongly from the Arrhenius behavior as the temperature is lowered towards T_g . The time dependence of the main (α) relaxation process in supercooled liquids is also not described by a simple exponential function, but can often be well approximated in the time domain by a stretched exponential function,

$$\Phi(t) = \exp[-(t/\tau)^\beta], \quad (1)$$

where β is called the stretching exponent, $0 < \beta \leq 1$, and the degree of non-exponentiality is given by $n = 1 - \beta$. These two properties seem to be connected, since n tends to be much smaller in strong glasses than in fragile ones.³ While such a connection is found in various model systems, these do not usually provide any simple explanation of it in terms of the microscopic properties of the system.

In this paper, we discuss a simple model system for which we find these properties, and also a simple explanation of them and their connection in terms of the average environments of the sites, namely the ferromagnetic q -spin Potts model⁴ of spins that can take any of q distinct values located on the sites of a lattice. At first sight, there seems to be very little resemblance between such a model of spins on a lattice and supercooled liquids (unless these spins are assumed to represent regions of the system with different properties, as in the facilitated kinetic Ising model of Fredrickson and Andersen,⁵ for instance). However, it is well known that the dielectric relaxation of plastic crystals such as ethanol is very similar to that of the corresponding supercooled liquid,⁶ so that spatial disorder is not an essential requirement for the typical behavior of supercooled liquids. For these plastic crystals, one widely-used model is that of thin rods rotating about their centres which are fixed on a lattice.⁷ An alternative model for these materials would be of rods rotating between a number of discrete orientations, which could be represented by spins as in the Potts model. In that case, the Potts model corresponds to a different (and in principle no less plausible) interaction between adjacent molecules, and so its properties may also be expected to resemble those of plastic crystals and supercooled liquids.

While the ferromagnetic Potts model has been extensively studied for over 50 years, most studies have been associated with its thermodynamic rather

than its relaxation properties. Even a recent study of the latter was concerned (like most recent papers) with a distribution of positive and negative values of the interaction J between equal spins on adjacent sites.⁸ The system that we study is the much simpler ordered ferromagnetic Potts model, in which the interaction J has the same positive value between any pair of equal spins on adjacent sites. A recent study of the dynamic properties of this ferromagnetic model⁹ was concerned with the dynamics of the phase transition rather than with relaxation some distance from it which we study. Our motivation for studying the Potts model were based on the following two observations about non-Arrhenius behavior and non-exponential relaxation. Firstly, both simple exponential relaxation and a fixed activation energy are associated with a fixed environment for the molecules, so that departures from both these types of behavior will occur if the environment of the molecules changes. Secondly, just as stretched exponential relaxation means that as time proceeds the relaxation rate decreases, so does the increase in the effective activation energy as the temperature is lowered mean that the relaxation rate is less than it would be for a fixed environment. In the ferromagnetic Potts model, as the temperature is lowered towards the phase transition temperature T_c , even before the critical region close to T_c is reached, larger and larger clusters of spins of the same size are expected to be formed, so that the environment of the spins changes to make transitions more difficult, as discussed in section 4. Similarly, at any temperature those spins that are inside a cluster of identical spins will have a lower probability of making a transition, so that as time proceeds increasing fractions of the spins that have not made a transition will be in such clusters, and this leads to a slowing down of the transition rate. Thus, the Potts model should provide a simple system in which, by tracking the changes in the environments of the sites, we can see a common source for non-Arrhenius behavior and non-exponential relaxation.

The idea of the importance of a molecule's local environment is not new, and has been considered previously by quite a number of authors. For instance, Ngai's coupling model essentially involves changes in the coupling of a molecule to its surroundings as time proceeds.¹⁰ The defect diffusion model of Bendler and his co-workers¹¹ can also be regarded as involving changes in the environment of free defects surrounding a molecule. In the theory of the fragility of supercooled liquids proposed by Tanaka,¹² the α -relaxation is associated with the dynamics of the formation and annihilation of solid-like islands, but he considers only its temperature dependence and not the time dependence of the relaxation. Such islands are examples of local inhomogeneities or heterogeneities, and these have been considered by Garrahan and his co-workers¹³ in systems with dynamic constraints. Numerous other approaches have been based on 2-state or 2-region models and dynamic constraints, such as the much studied Fredrickson-Andersen model.⁵ However, none of these approaches examined model systems with no externally imposed assumptions, such as the Potts model considered in this paper.

A detailed description of the q -spin ferromagnetic Potts model and its re-

laxation properties is presented in section 2, while section 3 contains the results of extensive computer simulations on it. These results are discussed and explained in section 4, and their implications for real supercooled liquids are then considered, while our conclusions are presented in section 5.

II. The ferromagnetic q -spin Potts model.

The Hamiltonian for the ordered ferromagnetic q -spin Potts model with interactions only between the spins at adjacent sites can be written as⁴

$$H = -J \sum_i \sum_{j(i)} \delta(\sigma_i, \sigma_j) \quad (2)$$

where $J > 0$, the first sum is over all the sites i in the system and the second one over all the sites $j(i)$ that are nearest neighbors of the site i , the spins σ_i can take any integer value between 1 and q , and δ is the Kronecker delta. Hence, the energy associated with a spin having z adjacent sites with the same spin is just $-zJ$. The probability of a change in the spin at a site which involves an increase of energy ΔE at temperature T was taken to have the standard form

$$\begin{aligned} w &= w_0, & \Delta E < 0 \\ &w_0 \exp(-\Delta E/k_B T), & \Delta E > 0 \end{aligned} \quad (3)$$

We note that the Potts model incorporates temperature explicitly, unlike models with dynamic constraints such as that of a system of rotating rods which has been applied to materials such as ethanol and in which the temperature had to be introduced in terms of the lengths of the rods.⁷

A question that always arises in simulations is whether to update the spins one at a time, which may be more realistic but requires a lot of computer time, or to update all of them simultaneously in one time step, which usually requires much less computer time but is not necessarily very realistic. For our model, which involves only interactions between spins on adjacent sites, we use the fact that the sites of the square lattice can be chosen as $\mathbf{n} = n_1\mathbf{a}_1 + n_2\mathbf{a}_2$, where $\mathbf{a}_1 = (1, 0)$ and $\mathbf{a}_2 = (0, 1)$. This lattice can be divided into four sub-lattices, with $(n_1, n_2) = (2j, 2k), (2j+1, 2k), (2j, 2k+1)$, and $(2j+1, 2k+1)$ where j and k are integers, such that adjacent sites are not on the same sub-lattice. Accordingly, the simultaneous updating of the spins on one sub-lattice at a time corresponds physically to updating the spins one at a time, but with a saving in computer time comparable to that obtained by updating all the spins at once. At each time step all the spins on one sub-lattice were updated simultaneously, while the sub-lattices were considered in turn. Because of possible problems with the Metropolis algorithm, in which the final state at each step is chosen at random, we used the equivalent of the continuous time Monte-Carlo algorithm,¹⁴ and considered at each step all possible transitions from each site with their appropriate probabilities.

The ferromagnetic q -spin Potts model on a square lattice for an infinite system exhibits a phase transition at a critical temperature T_c , and this transition is of second order for $q \leq 4$ and of first order for $q > 4$. In terms of the variable $v = \exp(J/[k_B T]) - 1$, the values of T_c are given by the solutions $v = v_c$ of the equation $v^2 = q$.¹⁵ For our simulations on a finite system we do not expect to observe an abrupt phase transition, and there are also considerable computational problems in reaching a steady state for temperatures too close to T_c , and so we restricted our calculations to $T_c/T \leq 0.99$. The quantity that we studied was the fraction $P(t)$ of sites at which the spin has not changed by time t . For convenience we call $P(t)$ the relaxation function, and the sites at which the spin has not changed as the unrelaxed sites. We considered $P(t)$, rather than a spin auto-correlation function such as $C(t, t') = \langle \sigma_i(t')\sigma_i(t + t') \rangle$ with $\sigma_i\sigma_j = \delta(\sigma_i, \sigma_j)$, because the latter contains a lot more random noise which will be strongly affected by the value of q . All the sites with no neighbors having the same spin were omitted from the calculation of $P(t)$, since for these $E = 0$ so that any change of spin has $\Delta E \leq 0$ and the relaxation on these sites would introduce into $P(t)$ a component that relaxed rapidly with time in a simple exponential manner. This component corresponds to a fast β -relaxation in a supercooled liquid, while the transitions that we consider resemble the much slower ones of the α -relaxation, in which we are interested here. The question of the possible merging of the α and β relaxations at high temperatures will be discussed elsewhere.

III. Results of the calculations

Extensive calculations were performed for $q = 3, 4, 5, 6, 8$ on a lattices of 200 by 200 sites with periodic boundary conditions, a size that was adequate to give very similar results for different sets of runs. For each value of q and T , an array of random spins was first annealed until the system's energy converged and then annealed further until the spins at 99% of the sites had changed. For high values of T_c/T , additional anneals were performed as necessary until a steady state was obtained. After this, five runs were performed on different initial states until $P(t)$ reached 0.01, and the average value of $P(t)$ was fitted to a stretched exponential function, equation (1). This fit was excellent not only on a double log plot, where many types of function appear as a straight line, but also on a graph of $\ln[P(t)]$ as a function of t , except for occasional slight deviations at very short or very long times. The results of the individual runs, which contained much more random noise, were also fitted to stretched exponential functions, and we checked that the average values of $\ln(\tau)$ and of β from the five runs were close to those obtained from fitting the average value of $P(t)$. Accordingly, all the results presented below relate to the values of the quantities from the average of five runs. Incidentally, the scatter in the results increased with increasing q , as is to be expected since the number of possible states in the system is 200^{2q} . It also increased with increasing $1/T$, and was much greater for β than for $\ln(\tau)$. For our calculations, it proved convenient to choose $w_0 = 0.5/(q - 1)$, so that

the maximum total transition probability at each step was the same, 0.5, for all the systems. Calculations were also performed for $q = 2$ (the usual Ising ferromagnet), but their results are somewhat different and so are not reported here. The reason for this difference, as discussed in section 4, is that for $q = 2$ any spins differing from those at a given site are equal to each other, and this affects the transitions rates for sites with $z = 2$.

In figure 1 we present the values of $\ln(\tau)$ as functions of $1/T$ for all the systems. In presenting the results, we choose $J = 1/k_B$, so that $J/(k_B T) = 1/T$. We only report here the results for $T_c/T \geq 0.5$, since this contains the temperature range of main interest for the α -relaxation. It is immediately apparent that the temperature dependence of $\ln(\tau)$ for all values of q differs considerably from an Arrhenius behavior, which would correspond to a straight line. While this temperature dependence does not correspond to any of the standard functions used for supercooled liquids,¹ it is qualitatively similar to that found for such liquids. We found that the results all fit well on to the master curve of $\ln(\tau)$ as a function of T_c/T shown in figure 2 (apart from very close to T_c), and so we show all our other results as a function of T_c/T rather than of $1/T$. The values of $J/(k_B T_c)$ are 1.005 for $q = 3$, 1.10 for $q = 4$, 1.174 for $q = 5$, 1.238 for $q = 6$, and 1.342 for $q = 8$. In figure 3, we show the stretching exponent β as a function of T_c/T , and note at once that (at least for $T_c/T \leq 0.95$) the values of β decrease steadily as T_c/T increases (and so the temperature decreases).

As we discuss in the next section, the interpretation of these results is connected with changes in the environments of the sites, and so we now present the results of our calculations on them. We found two useful ways of describing these environments. The first is in terms of the average value $\langle z \rangle$ of the number z of adjacent sites having the same spin as that of the given site, which we refer to as the spin correlation. It is convenient to divide $\langle z \rangle$ by the coordination number, $z_c = 4$, so that its maximum value is unity, and we call the resulting quantity the reduced spin correlation sc . The other quantity that we examined is the fraction of sites inside clusters of the same spin (and not on their boundaries), so that the spin on the site is the same as that on all its adjacent sites, and we denote this quantity by cl . Since the sites in clusters all contribute unity to sc , the difference $sc - cl$ is associated with sites on the edges of clusters or outside them. The importance of the environment for the relaxation rates is associated with its relationship to the probability for a change of spin, as discussed in the next section.

The major properties that are of interest with regard to the environments of the sites are how these change with temperature and with time. In order to show the change in the environment as a function of time, we present in figures 4 and 5 sc and cl as functions of the fraction P of unrelaxed sites for $T_c/T = 0.9$ for all five values of q . As an example of how this change in time varies with temperature, we present in figure 6 cl as a function of P for $q = 5$ and three different values of T_c/T , 0.7, 0.8 and 0.9, for which the values of the stretching exponent β were 0.86, 0.79 and 0.72 respectively. In these figures, we chose P to decrease from 1 to 0 on the x -axis, as this is the direction of increasing time.

Finally, in order to show the change in environment as a function of temperature, we show in figures 7 and 8 the steady state values of sc and cl as functions of T_c/T .

IV. Discussion

A comparison of figures 1-3 with figures 4-8 shows clearly that both non-exponential relaxation and non-Arrhenius temperature dependence are associated with the change in the average environment of the sites with temperature and time, as we suggested in the Introduction. We now consider in turn the temperature dependence and dependence on q of the environments of the sites, the relationship between the transition rates and the environments, and the time dependence of the environments. After this, we discuss the implications of our results for understanding the properties of supercooled liquids.

The departure from an Arrhenius law of the temperature dependence of $\ln(\tau)$ is associated with the increase as the temperature is lowered of the reduced spin correlation sc and of the fraction of states inside clusters cl . The reason for these increases is that the energy associated with a site decreases as its spin correlation number z increases, while the number of possible arrangements of the spins associated with such a state decreases, which leads to a decrease in the system's entropy. As the temperature is lowered the role of entropy in the free energy becomes less important, and this leads to an increase in $\langle z \rangle$ and so in sc , as well as to an increase in the fraction cl of sites within clusters where their energy is lowest. In order to obtain a better qualitative understanding of this temperature dependence, let us suppose that the environments of the different sites could all be chosen independently. If $C_1(z)$ denotes the number of possible arrangements of spins round a given site, then the single site partition function would be

$$Z_1(T) = \sum_z C_1(z) \exp[zJ/(k_B T)], \quad (4)$$

and the fraction of sites with a given value of z would be

$$n_1(z, T) = C_1(z) \exp[zJ/(k_B T)]/Z_1(T). \quad (5)$$

If the sites with $z = 0$ are included in the sum in equation (4), it can readily be shown that for the square lattice $Z_1(T) = \{q - 1 + \exp[J/(k_B T)]\}^4$. The fraction of states in a cluster in this approximation is $1/Z_1(T)$, which increases as T decreases and decreases as q increases. Since the arrangement of spins around adjacent sites is obviously not independent, the entropy of the system cannot be calculated just in terms of these single site quantities $C_1(z)$, but equations (4)-(5) are useful for qualitative arguments such as the above. In particular, they explain not only the increase in the fraction cl of particles in clusters as the temperature is lowered, as shown in figures 6 and 8, and the increase in the reduced spin correlation function sc shown in figure 7, but also why an increase in the number of possible values q of the spin on a site leads to a reduction in the values of cl and sc , as found in figures 4, 5, 7 and 8.

We now turn to the relationship between the environment of the sites as characterized by the functions sc and cl and the transition rates. For sites inside a cluster, any transition reduces z from z_c to zero, and so requires the maximum possible activation energy $E_{act} = z_c J = 4J$, so that cl describes the fraction of sites at which a transition requires this activation energy. For a site with $z = 3$, there is one transition that reduces z from 3 to 1 and so requires an activation energy of $2J$, while any other transition requires an activation energy of $3J$. For sites with $z \leq 1$ transitions are possible that do not require an increase in energy, while for $z = 2$ such transitions are only possible if the neighboring sites contain two pairs of equal spins. Incidentally, this is always the case for $q = 2$, which is why the results for this value of q differ somewhat from those for $q > 2$ and so we decided not to report them here. For the sites with values of z that require thermal activation for the transitions, an alternative relaxation mechanism is that the value of z at that site decreases as a result in changes in the spin at adjacent sites until unactivated transitions (or ones with a lower activation energy) are possible. In order to find out which process is actually the dominant one, we performed a number of calculations of the fraction of sites initially having different values of z , the fraction at which the first transition occurs for a given value of z , and the fraction at which the first transition involves a given activation energy. In a typical set of results, for $q = 5$ and $T_c/T = 0.9$, we found that 11% of the sites have $z = 0$, 24% have $z = 1$, 30% have $z = 2$, 24% have $z = 3$, and 11% have $z = 4$. During the relaxation process we found that the neighborhoods of the unrelaxed sites change, since at only 2% of the sites did the first transition occur with $z = 4$, i.e. at sites within a cluster, while at 10% it occurred when $z = 3$, at 34% when $z = 2$, at 38% when $z = 1$, and at 16% when $z = 0$. A comparison of these results with the values of z for the whole system shows that the dominant mechanism for the relaxation of sites with high spin coordination number z is for the spins on some of the adjacent sites to change before they relax, so as to reduce the activation energy required for a transition. This is especially clear for sites initially within a cluster (for which $z = 4$), since 9/11 of these sites had lower values of z when the first transition took place. As a result of these changes of environment, 55% of the first transitions did not require any activation energy, 24% required an activation energy of J , 14% an activation energy of $2J$, 5% an activation energy of $3J$, and 2% an activation energy of $4J$. Such a behavior was found for all of the values of T_c/T that we studied (up to $T_c/T = 0.99$). The time required for this process is longest for sites within clusters, and increases with increasing cluster size, while the average cluster size increases with increasing cl , and so as the temperature decreases. This is one of the main sources of the departure from an Arrhenius law temperature dependence of the mean relaxation time.

With regard to the time dependence of the relaxation, those sites with a lower initial value of z will have the fastest relaxation rates and so tend to relax first, while relaxation at the other sites will take longer as it requires either a larger activation energy or a change in their value of z as a result of transitions of their neighbors. As a result, sc and cl for the unrelaxed sites increases with

time, as shown in figures 4-6, and this leads to the observed departure from simple exponential relaxation. As can be seen from figure 6, as T_c/T increases the change in the environment (as measured by cl) increases, and this correlates with the decrease in β , which reinforces our conclusion that the non-exponential time dependence is associated with changes in the environment of the sites.

We now turn to some of the implications of our results for real supercooled liquids. Our results strongly suggest that in real supercooled liquids the temperature and time dependence of the relaxation are both associated with changes in the average environments of the molecules, which explains why they are often connected. This implies that a key factor in determining the properties of supercooled liquids is the number of different environments available for the molecules. Since this is smallest for covalently bonded materials such as SiO_2 , this explains why these are the strongest liquids, with the smallest deviations from an Arrhenius law temperature dependence of their relaxation times and from a simple exponential time dependence of their relaxations. On the other hand, there are numerous possible molecular environments for van-der-Waals bonded liquids, which explains why these tend to be the most fragile and to have the lowest stretching exponents for their relaxation. H-bonded liquids have an intermediate number of possible molecular environments, because of the restrictions imposed by the hydrogen bonds, and so their fragility and stretching exponents tend to be intermediate between those of covalently bonded and van-der-Waals bonded liquids. In addition, since the glass transition involves a freezing of the molecular environments, the decrease in entropy when this happens will increase with the number of possible environments, and so with the fragility, as is found experimentally. A more detailed analysis of these effects is beyond the scope of this paper. Our results also explain qualitatively the connection between fragility and the degree of non-exponential relaxation observed in many supercooled liquids.³ The effects of changes in the environment on the lifetimes of dynamic heterogeneities found in supercooled liquids is most easily discussed if one associates these heterogeneities with molecules inside clusters, which are often assumed to be solid-like. These clusters are the states of lowest local energy, so that an increasing fraction of molecules occupy them as the temperature is lowered. In principle, just as for the Potts model, such molecules can relax either by a particle in the middle of a cluster acquiring enough activation energy to change its state or particles on the edge of a cluster relaxing in turn so that the clusters of unrelaxed particles gradually disintegrate. These two processes resemble, respectively, the formation and motion of interstitial atoms in a crystal and the dissociation of subcritical nuclei in a liquid. If the latter process is more probable, as one would expect at temperatures above the glass transition temperature just as it was found to be for the Potts model at temperatures above T_c , then the lifetimes of the dynamic heterogeneities will be of the same order of magnitude as the α -relaxation time, as is found experimentally in many liquids^{16,17}.

V. Conclusions

One major advantage of the Potts model studied in this paper over many other models is that it does not require the imposition of any external assumptions, such as the existence of liquid-like and solid-like regions or of dynamic restrictions, although these may well be present in the model. Instead, all the results are derived directly from the system's Hamiltonian. The results of our calculations on the two-dimensional ferromagnetic Potts model show that both the temperature dependence and the time dependence of the relaxation are associated with changes in the arrangements of spins around the different sites, i.e. the environments of the sites. Such a feature can explain why covalently bonded liquids, in which there is only a small number of possible molecular environments, are strong, while van-der-Waals bonded liquids, in which this number is large, tend to be the most fragile. It also explains the connection between stretched exponential relaxation and a non-Arrhenius temperature dependence of the relaxation time in supercooled liquids. In addition, the relaxation in the Potts model of spins within clusters of sites by the disintegration of the clusters suggests that a similar mechanism is relevant to the relaxation of molecules inside dynamic heterogeneities, and this would explain why the lifetimes of such heterogeneities is usually found to be of the same order of magnitude as the α -relaxation time.

References

- ¹C. A. Angell, K. L. Ngai, G. B. McKenna, P. F. McMillan, S. W. Martin, J. Appl. Phys. **88**, 3114 (2000)
- ²C. A. Angell, J. Phys. Chem. Solids **49**, 863 (1988)
- ³K.L. Ngai, J. Non-Cryst. Sol. **275**, 7 (2000)
- ⁴F. Y. Wu, Rev. Mod. Phys. **54** 235 (1982)
- ⁵G.H. Fredrickson and H.C. Andersen, J. Chem.Phys. **83** 5822 (1985)
- ⁶S Benkhof, A Kudlik, T Blochowicz and E Rossler, J. Phys: Condens. Matter **10** 8155 (1998)
- ⁷F J Bermejo, M Jimenez-Ruiz, A Criado, G J Cuello, C Cabrillo, F R Trouw, R Fernandez-Perea, H Lowen and H E Fischer, J. Phys: Condens. Matter **12** A391 (2000)
- ⁸C Brangian, W Kob and K Binder, J. Phys. A: Math. Gen. **36** 10847 (2003)
- ⁹A. Velytsky, B. A. Berga and U. M. Hellera, Nuclear Physics B (Proc. Suppl.) **119** 861 (2003)
- ¹⁰K L Ngai, Comments Solid State Phys. **9** 121 (1979)
- ¹¹J. T. Bendler, J. J. Fontanella and M. F. Shlesinger, J. Chem. Phys. **118** 6713 (2003)
- ¹²H Tanaka, J. Non-Cryst. Sol. **351** 3385 (2005)
- ¹³J. P. Garrahan and D. Chandler, Phys. Rev. Lett. **89**.035704 (2002); Berthier L and Garrahan J P, J. Chem. Phys. **119** 4367 (2003).
- ¹⁴V. Gotcheva, Y. Wang, A. T. J. Wang and S. Teitel, Phys. Rev. B **72**, 064505 (2005).
- ¹⁵R. J. Baxter, J. Phys. C: Solid State Phys. **6** L445 (1973).
- ¹⁶E V Russell and N E Israeloff, Nature **408** 695 (2000)
- ¹⁷H Sillescu, R Bohmer, G. Diezemann and G. Hinze, J. Non-Cryst. Sol. **307-310** 16 (2002)

Figure captions.

Figure 1.(Color on line) The relaxation time τ of the stretched exponential relaxation of $P(t)$ as a function of $1/T$ for $q = 3$ (circles, red), $q = 4$ (stars, violet), $q = 5$ (squares, black), $q = 6$ (hexagons, magenta) and $q = 8$ (triangles, blue). The dotted lines are just to guide the eye.

Figure 2. (Color on line) The relaxation time τ of the stretched exponential relaxation of $P(t)$ as a function of T_c/T for $q = 3$ (circles, red), $q = 4$ (stars, violet), $q = 5$ (squares, black), $q = 6$ (hexagons, magenta) and $q = 8$ (triangles, blue). The dotted lines are just to guide the eye.

Figure 3. (Color on line) The stretching exponent β of the stretched exponential relaxation of $P(t)$ as a function of T_c/T for $q = 3$ (circles, red), $q = 4$ (stars, violet), $q = 5$ (squares, black), $q = 6$ (hexagons, magenta) and $q = 8$ (triangles, blue). The dotted lines are just to guide the eye.

Figure 4. (Color on line) The reduced correlation function $sc(P)$ as a function of P for $T_c/T = 0.9$ for $q = 3$ (red), $q = 4$ (violet), $q = 5$ (black), $q = 6$ (magenta) and $q = 8$ (blue).. The values of q are shown on the curves.

Figure 5. (Color on line) The fraction of sites in clusters $cl(P)$ as a function of P for $T_c/T = 0.9$ for $q = 3$ (red), $q = 4$ (violet), $q = 5$ (black), $q = 6$ (magenta) and $q = 8$ (blue).. The values of q are shown on the curves.

Figure 6. The fraction of sites in clusters $cl(P)$ for $q = 5$ as a function of P for $T_c/T = 0.7$.(dashes, blue), $T_c/T = 0.8$.(dot-dash, red) and $T_c/T = 0.9$.(solid, black). The values of T_c/T are marked on the curves.

Figure 7. (Color on line) The steady state reduced correlation function sc_0 as a function of T_c/T for $q = 3$ (circles, red), the top curve, $q = 4$ (stars, violet), $q = 5$ (squares, black), $q = 6$ (hexagons, magenta) and $q = 8$ (triangles, blue), the bottom curve. The dotted lines are just to guide the eye.

Figure 8. (Color on line) The steady state fraction of sites in clusters cl_0 as a function of T_c/T for $q = 3$ (circles, red), the top curve, $q = 4$ (stars, violet), $q = 5$ (squares, black), $q = 6$ (hexagons, magenta) and $q = 8$ (triangles, blue), the bottom curve. The dotted lines are just to guide the eye.















